Integral light-scattering and absorption characteristics of large, nonspherical particles

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We obtain and analyze simple analytical formulas for asymmetry parameters and absorption cross sections of large, nonspherical particles. The formulas are based on the asymptotic properties of these characteristics at strong and weak absorption of radiation inside particles. The absorption cross section depends on parameter ϕ, which determines the value of the light-absorption cross section for weakly absorbing particles. It is larger for nonspherical scatterers. The asymmetry parameter depends on two parameters. The first is the asymmetry parameter g_0 of a nonspherical, transparent particle with the same shape as an absorbing one. The second parameter, β, determines the strength of the influence of light absorption on the value of the asymmetry parameter. Parameter β is larger for nonspherical particles. One can find these three parameters (ϕ, g_0, and β) using a ray-tracing code (RTC) for nonabsorbing and weakly absorbing particles. The RTC can then be used to check the accuracy of the equations at any absorption for hexagonal cylinders and spheroids. It is found that the error of computing the absorption cross section and $1 - g$ (g is the asymmetry parameter) is less than 20% at the refractive index of particles $n = 1.333$. Values for asymmetry parameters of large, nonabsorbing, spheroidal particles with different aspect ratios are tabulated for the first time to our knowledge. They do not depend on the size of particles and can serve as an independent check of the accuracy of T-matrix codes for large parameters. © 1997 Optical Society of America

1. Introduction

Most particulate natural media (ocean water, dust aerosols, snow, cirrus clouds, and foam) are composed of nonspherical particles. One can study optical properties of such scatterers by solving Maxwell's equations.[1–4] Unfortunately, this approach is characterized by poor numerical stability for nonspherical particles that are large compared with wavelength. One can avoid this problem by using the geometrical-optics approximation (GOA).[5–10] The accuracy of this approximation for spheroidal particles can be studied using an improved version of the exact T-matrix method as described in Ref. 11. Research in this area has already begun.[12] It was reported in Ref. 12 that the error of phase-function calculation for spheroids using the GOA is less than that for surface-equivalent, spherical particles. The GOA gives sufficiently accurate phase functions for spheroids with size parameters exceeding 60.[12]

Here we investigate integral light-scattering characteristics (extinction cross section C_{ext}, absorption cross section C_{abs}, light-pressure cross section C_{pr}, and asymmetry parameter g) of large, nonspherical particles. These values are of fundamental importance to the radiative transfer theory.[13] For example, according to the similarity principle,[13,14] particulate media with different phase functions but the same parameters $[1 - \omega^* = (1 - \omega)/(1 - \omega g)$ and $\tau^* = (1 - \omega g)\tau]$ have approximately the same radiative characteristics. Here $\omega = 1 - C_{\text{abs}}/C_{\text{ext}}$ is the single-scattering albedo, and τ is the optical thickness.

Note that phase functions and especially polarization characteristics of spherical and volume- or surface-equivalent, nonspherical particles are considerably different.[15–17] However, integral light-scattering characteristics of nonspherical particles can be represented in many cases by those of effective spheres. For example, the extinction cross section C_{ext} of a large particle ($\rho = 2\pi a/\lambda \gg 1$, $2\rho|m - 1| \gg 1$, where a is the size of the particle, λ is the wave-

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Received 20 June 1996; revised manuscript received 11 March 1997.
0003-6935/97/338785-06$10.00/0

length, and $m = n - i\chi$ is the refractive index of the particle) is given by the following equation[1,2]:

$$C_{\text{ext}} = 2S, \tag{1}$$

where S is the projected area of the particle. Thus spherical and nonspherical large particles with the same values of S have the same extinction cross sections. Note that the average cross section $\langle S \rangle$ is equal to $\Sigma/4$ and $C_{\text{ext}} = \Sigma/2$ for randomly oriented, convex particles.[2] Here Σ is the particle's surface area.

In many applications[2,13] it is important to know not only C_{ext} but also the particles' asymmetry parameters g, single-scattering albedos ω, absorption cross sections C_{abs}, and light-pressure cross sections C_{pr}.

There are many simple analytical solutions for the absorption cross section of nonspherical particles (see Refs. 6, 8, 18, and 19). However, their application is limited by special cases such as small values of the real or imaginary part of the refractive index of particles. Parameterizations for the asymmetry parameters and light-pressure cross sections of nonspherical particles are unknown to us. Thus here we propose simple approximate formulas for calculating integral light-scattering characteristics (g, C_{abs}, ω, C_{pr}) of large, nonspherical, convex particles within the framework of the GOA. With this in mind, we use asymptotic properties of the absorption cross section and the asymmetry parameter for weak and strong absorption.

To simplify the problem, we assume that $\chi \ll n$. This condition holds for most substances in the optical range of the spectrum. The product $\chi\rho$ can be arbitrarily large.

2. Physical Parameterization

A. Absorption Cross Section

To obtain an approximate solution for the absorption cross section, we used the following premises:

(1) The absorption cross section of a large, weakly absorbing ($c = 4\chi\rho \to 0$), nonspherical particle is proportional to the volume of a particle V and the absorption coefficient $\alpha = 4\pi\chi/\lambda$. Thus it follows at $c \ll 1$ that[3,6–8]:

$$C_{\text{abs}} = \phi(n, \xi)\alpha V, \tag{2}$$

where the coefficient $\phi(n, \xi)$ depends on the real part of the refractive index of a particle and its shape. The shape is characterized by the vector parameter ξ. For spherical particles it follows that[3,10]

$$\phi(n, \xi) = n^2(1 - b^3), \tag{3}$$

where $b = (1 - 1/n^2)^{1/2}$.

(2) One can determine the absorption cross section of a large, strongly absorbing particle ($c \to \infty$) by the following equation[3,10]:

$$C_{\text{abs}} = [1 - R(n, \xi)]S, \tag{4}$$

Table 1. Dependence of Values *R* and g_∞ on the Refractive Index of Particles *n*

n	R	g_∞
1.1	0.0252	0.9946
1.2	0.0443	0.9856
1.333	0.0664	0.9714
1.4	0.0768	0.9638
1.5	0.0918	0.9520
1.6	0.1063	0.9400
1.7	0.1203	0.9280

where $R(n, \xi)$ is part of the incident energy reflected by a particle. It would appear to be reasonable that the value of C_{abs} at any value of c can be represented by

$$C_{\text{abs}} = [1 - R(n, \xi)]\{1 - \exp[-\psi(n, \xi)c]\}S, \tag{5}$$

where $c = 4\chi\rho$, $\rho = 2\pi a/\lambda$, $a = 3V/4S$, and

$$\psi(n, \xi) = \frac{2\phi(n, \xi)}{3[1 - R(n, \xi)]}. \tag{6}$$

The value of a is equal to the radius of a particle (for spheres). Note that Eq. (5) has correct limits at $c \to 0, \infty$ [see Eqs. (2) and (4)]. The accuracy of Eq. 5 is checked for spheroidal and hexagonal particles in Section 3.

For randomly oriented, convex, nonspherical particles $\langle R(n, \xi)\rangle$ is equal to $R(n)$,[2,20] where $R(n)$ is the part of the energy reflected by a spherical particle (see Table 1[10]) and $\langle \ldots \rangle$ means averaging on orientations. Thus it follows approximately from Eq. (5) for randomly oriented, monodispersed, nonspherical particles that

$$\langle C_{\text{abs}} \rangle = \frac{1}{4}[1 - R(n)]\{1 - \exp[-\psi(n, \xi)c]\}\langle \Sigma \rangle, \tag{7}$$

where $c = 4\chi\rho$, $\rho = 2\pi a/\lambda$, and $a = 3V/\Sigma$. One can find equations for volumes V and surface areas Σ of different nonspherical particles in Refs. 8 and 21 (see Table 2).

The absorption cross section in Eq. (7) depends on

Table 2. Surface Areas and Volumes of Spheroids and Hexagonal Cylinders[a]

Type of Particle	Surface Area	Volume
Oblate spheroid	$2\pi a_1^2\left(1 + \frac{\xi^2}{2\delta}\ln\frac{1+\delta}{1-\delta}\right)$	$\frac{4\pi}{3}a_1^2 a_3$
Prolate spheroid	$2\pi a_1^2\left(1 + \frac{\xi}{\delta}\arcsin\delta\right)$	$\frac{4\pi}{3}a_1^2 a_3$
Hexagonal cylinder	$3l^2\left(\sqrt{3} + 2\frac{L}{l}\right)$	$\frac{3\sqrt{3}}{2}Ll^2$

[a] a_1, a_2, and a_3, are semiaxes of a spheroid, $\xi = a_3/a_1$, for an oblate spheroid $\delta = (1 - \xi^2)^{1/2}$, and for a prolate spheroid $\delta = (1/\xi)(\xi^2 - 1)^{1/2}$. L is the length of a hexagonal cylinder, and l is the side of a hexagonal cylinder's cross section.

Table 3. Value of g_0 for Randomly Oriented, Spheroidal Particles at Different Values of Shape Parameter ξ and the Real Part of Refractive Index n

n/ξ	0.3	0.5	0.7	1.0	1.5	2.0	3.5	∞
1.1	0.9582	0.9583	0.9764	0.9731	0.9652	0.9642	0.9711	0.9817
1.2	0.9123	0.8976	0.9127	0.9341	0.9154	0.9158	0.9413	0.9551
1.333	0.8774	0.8129	0.8428	0.8843	0.8510	0.8556	0.9085	0.9201
1.4	0.8590	0.7718	0.8041	0.8613	0.8208	0.8298	0.8911	0.9029
1.5	0.8257	0.7135	0.7522	0.8299	0.7747	0.7965	0.8624	0.8795
1.6	0.7873	0.6677	0.7065	0.8015	0.7322	0.7685	0.8320	0.8577
1.7	0.7518	0.6407	0.6665	0.7759	0.6934	0.7479	0.8020	0.8360

parameter ψ only. It can be determined from C_{abs} at small values of c [see Eqs. (2) and (6)]:

$$\psi(n, \xi) = \frac{2\langle C_{\mathrm{abs}}\rangle}{3\alpha V[1 - R(n)]}. \tag{8}$$

To find ψ, one should calculate the value of $\langle C_{\mathrm{abs}}\rangle$ for weakly absorbing particles of different shapes using a ray-tracing code (RTC).[5,8,9] Note that the value of ψ does not depend on the size of particles and the imaginary part of the refractive index. Here we consider only spheroids and hexagonal particles and use the RTC[5] for computations of this value.

B. Light-Pressure Cross Section and Asymmetry Parameter

The light-pressure cross section C_{pr} is defined as follows[2]:

$$C_{\mathrm{pr}} = (1 - g)C_{\mathrm{ext}} + gC_{\mathrm{abs}}. \tag{9}$$

Thus, to calculate the light-pressure cross section, one has to know the value of the asymmetry parameter g along with the values of C_{ext} and C_{abs} [see Eqs. (1) and (5)]. To find the asymmetry parameter, one must use the following premises:

(1) The asymmetry parameter for a nonabsorbing (g_0) or strongly absorbing (g_∞) large particle does not depend on the particle size[2] and the value of the imaginary part of refractive index $\chi \ll n$ but only on its shape and the real part of refractive index n.

(2) For spherical particles it follows that[10]

$$g = g_\infty(n) - [g_\infty(n) - g_0(n)]\exp[-\beta(n)c], \tag{10}$$

where values of g_∞, g_0, and β depend on the real part of the refractive index only.

It is reasonable to suppose that Eq. (10) can be applied to large, nonspherical particles as well. In this case values of g_∞, g_0, and β depend on a specific particle shape and the real part of refractive index n. The dependence of these parameters on the size of particles and the imaginary part of the refractive index can be neglected.

For randomly oriented particles $\langle g_\infty(n, \xi)\rangle$ is equal to $g_\infty(n)$ for spheres[2,10,20] (see Table 1), and the value of the asymmetry parameter depends only on two parameters (g_0 and β). They can be found with the use of RTC's[5,8,9] by computing the asymmetry parameters g of particles with different shapes at $c = 0$ ($g_0 = g$) and $c \to 0$ $\{\beta = (g - g_0)/[(g_\infty - g_0)c]\}$.

3. Accuracy of Approximations

Here we find parameters g_0, β, and ϕ [ϕ determines ψ in Eq. (7)] for nonspherical particles in random orientation at different values of n and ξ, and we investigate the applicability and accuracy of Eqs. (7) and (10) for monodispersed, randomly oriented, spheroidal and hexagonal particles. Note that these parameters do not depend on the imaginary part of the complex refractive index or on particle size.

It is difficult to find the analytical solutions for these functions. Therefore we calculated asymmetry parameters and absorption cross sections of large, randomly oriented spheroids to obtain these functions numerically using the RTC developed by Macke *et al.*[5] for the following input parameters: $n = 1.1$, 1.2, 1.333, 1.5, 1.6, 1.7; $\lambda = 2\pi$; $a_1 = a_2 = 1000$; $a_3 = \xi a_1$, $\xi = 0.3, 0.5, 0.7, 1.0, 1.5, 2.0, 3.5, 5.0$; $\chi = 0$ (to find g_0); and $\chi = 10^{-5}$ (to find β and ϕ). Here a_1, a_2, and a_3 are the semiaxes of a spheroid. Note that $\xi < 1$ for oblate spheroids, $\xi = 1$ for spheres, and $\xi > 1$ for prolate spheroids.

Results of the computation of asymmetry parame-

Table 4. Values of $\beta(n, \xi)$ for Randomly Oriented, Spheroidal Particles at Different Values of Shape Parameter ξ and the Real Part of Refractive Index n

n/ξ	0.3	0.5	0.7	1.0	1.5	2.0	3.5
1.1	2.49	1.32	0.90	0.47	0.86	1.04	1.43
1.2	2.21	1.59	0.83	0.54	1.05	1.10	1.37
1.333	3.09	1.59	1.16	0.76	1.09	1.27	1.40
1.4	3.21	1.66	1.26	0.82	1.13	1.32	1.48
1.5	3.53	1.73	1.29	0.83	1.18	1.32	1.65
1.6	3.70	1.78	1.37	0.86	1.23	1.35	1.83
1.7	3.83	1.84	1.43	0.89	1.28	1.40	1.98

Table 5. Values of $\phi(n, \xi)$ for Randomly Oriented, Spheroidal Particles at Different Values of Shape Parameter ξ and the Real Part of Refractive Index n

n/ξ	0.3	0.5	0.7	1.0	1.5	2.0	3.5
1.1	2.46	1.66	1.32	1.11	1.14	1.18	1.24
1.2	2.78	1.84	1.45	1.18	1.25	1.29	1.37
1.333	3.16	2.09	1.60	1.24	1.36	1.43	1.52
1.4	3.35	2.21	1.67	1.26	1.42	1.49	1.59
1.5	3.64	2.40	1.78	1.29	1.50	1.58	1.70
1.6	3.93	2.59	1.88	1.31	1.57	1.68	1.80
1.7	4.21	2.76	2.00	1.33	1.64	1.76	1.89

ters g_0, β, and ϕ at different values of ξ and n are given in Tables 3–5 and Figs. 1–3.

For $\xi \in (0.3, 1.0)$ and $\xi \in (1.0, 2.5)$, g_0 for spheroids is less than it is for spheres ($\xi = 1$). As expected, $g_0 \to 1$ at $\xi \to 0$ and $g_0 \to g_c$ at $\xi \to \infty$, where g_c is the asymmetry parameter of nonabsorbing, infinite, circular cylinders with random orientation.[17] Data for the value of g_c are shown in the last column of Table 3. The data are larger than the values of g_0 for spherical particles.

Results for β are given in Table 4 and Fig. 2. Note that this function has a minimum at $\xi = 1$, which implies that the influence of absorption on the asymmetry parameter (and thus on phase function) is more important for nonspherical particles than for spheres. Interestingly, the function $\phi(n, \xi)$ also has a minimum at $\xi = 1$ (see Fig. 3 and Table 4). Thus weakly absorbing spheres absorb less energy than volume-equivalent, weakly absorbing, spheroidal particles [see Eq. (2)]. This is most likely because spheres have minimal surface area compared with nonspherical particles of the same volume.

Thus all auxiliary functions in Eqs. (7) and (10) were determined. Let us study the applicability and the accuracy of these equations in calculating the integral light-scattering characteristics of large, randomly oriented, spheroidal particles at any absorption. To this end, we calculated values of the asymmetry parameter and the absorption and light-pressure cross sections using the RTC[5] and Eqs. (7) and (10) at $n = 1.333$, $\chi = 10^{-p}$, $p = 2, 3, 4, 5, 6$, $a = b = 1000$, and $\xi = 0.5, 2.0$.

We studied the accuracy of approximations for the probability of photon absorption $1 - \omega$ and the similarity parameter[13] $s = [(1 - \omega)/(1 - g\omega)]^{1/2}$ as well. The results, given in Figs. 4(a) and 4(b), show that the error of the proposed approximate formulas is less than 10% in this case. Thus the proposed equations indeed can be applied for calculating integral light-scattering characteristics of nonspherical particles. Note that parameters g_0, β, and ϕ at intermediate values of n and ξ can be found by interpolation. Such interpolation is rather accurate at $\xi \geq 0.5$. However, at $\xi < 0.5$ functions $\beta(\xi)$ and $\phi(\xi)$ change rapidly, and more points are needed for accurate interpolation (not just $\xi = 0.3$ as in Tables 3–5).

As noted, Eqs. (7) and (10) are not limited to spheroids but can be applied to particles of different shapes (such as ellipsoids and circular and hexagonal

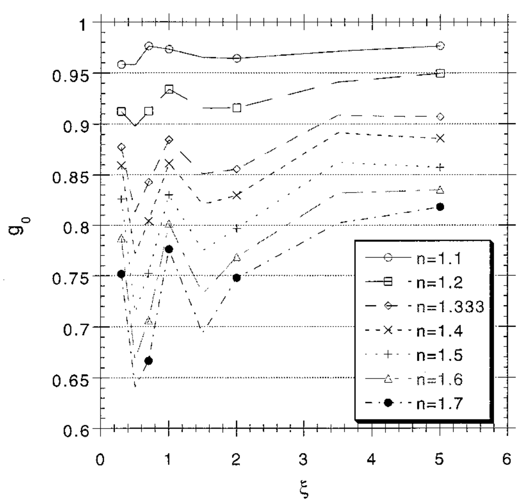

Fig. 1. Dependence of asymmetry parameter g_0 of nonabsorbing spheroids on the value of ξ at different values of the refractive index of particles n.

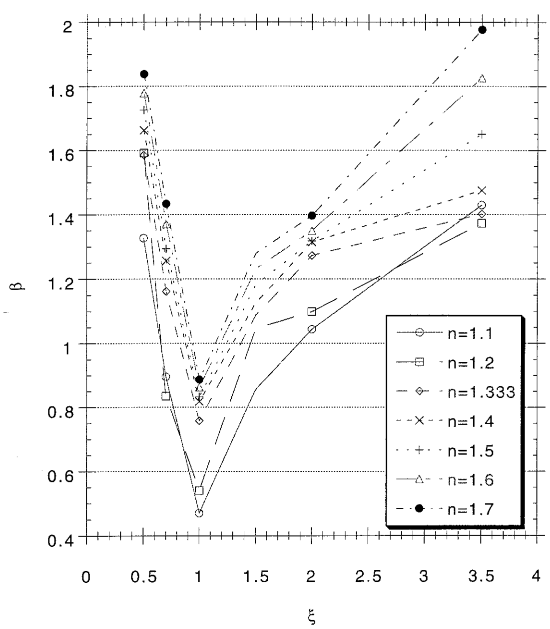

Fig. 2. Dependence of function β on the value of ξ at different values of the refractive index of particles n.

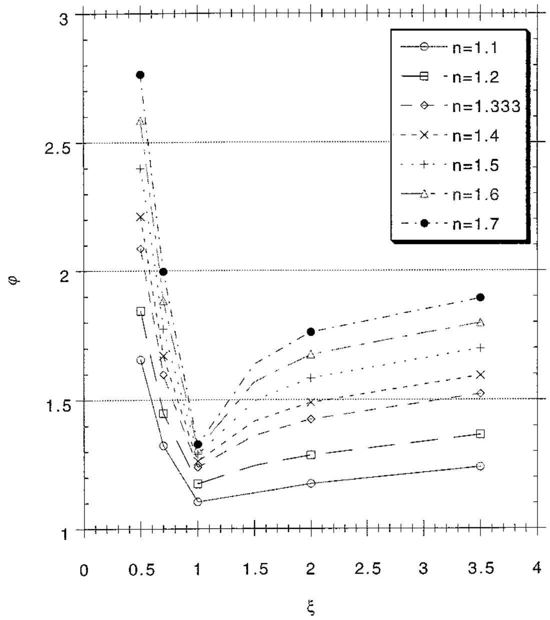

Fig. 3. Dependence of function ϕ on the value of ξ at different values of the refractive index of particles n.

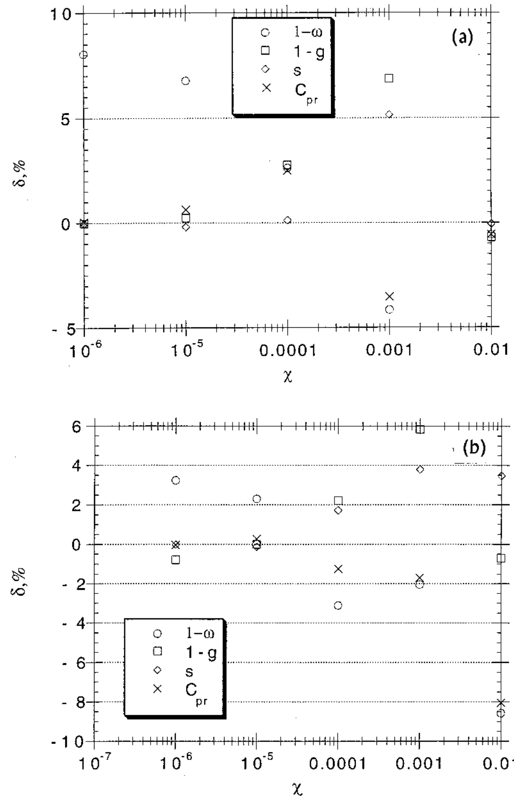

Fig. 4. Dependence of the relative error of approximate formulas for computing $1 - \omega$, $1 - g$, s, and C_{pr} at $n = 1.333$, (a) $\xi = 0.5$ and (b) $\xi = 2.0$, on the value of the imaginary part of the refractive index χ.

cylinders). To apply these formulas, one should first calculate g_0, β, and ϕ.

For example, the value of a in Eqs. (7) and (10) for hexagonal cylinders can be determined by the following equation (see Table 2):

$$a = \frac{L}{2 + 2L/s}, \tag{11}$$

where $s = \sqrt{3}l/2$, l is the side of a hexagon's cross section, and L is its length. Values of $g_0(n, \xi)$, $\beta(n, \xi)$, and $\phi(n, \xi)$ in Eqs. (7) and (10) were calculated using the RTC[5] for hexagons at $n = 1.333$ and are given in Table 6. We found that the errors in Eqs. (7) and (10) for hexagonal cylinders were less than 20% at $n = 1.333$, $\zeta \in (0.25, 10)$, and $\chi = 10^{-6} - 10^{-2}$. Here $\zeta = L/D$, where $D = \sqrt{3}l$ is the distance between opposite sides of a hexagonal cylinder.

4. Conclusion

We have obtained and analyzed approximate equations for absorption cross sections and asymmetry parameters of large, nonspherical particles. The equations can be used to investigate the dependence of the radiative characteristics of light-scattering media composed of large, nonspherical particles on their microstructure parameters. We studied the applicability of these equations for randomly oriented spheroids and hexagonal cylinders at different values of the imaginary part of the refractive index of particles and shape parameters. The error was found to be less than 10% for $1 - g$ and C_{abs} at $n = 1.333$ for spheroidal particles; it is less than 20% for hexagonal cylinders. A broader coverage of refractive indices, including the refractive index of ice in visible and infrared bands, will be the subject of a future study.

Absorption cross sections of large, weakly absorbing, nonspherical particles, such as ice crystals in clouds and snow, are larger than those for volume-equivalent spheres. Thus the single-scattering albedo $\omega = 1 - C_{\mathrm{abs}}/C_{\mathrm{ext}}$ for weakly absorbing, nonspherical particles is larger than that for spheres with the same effective radius $a = 3V/\Sigma$.

Values of g_∞ for convex, nonspherical particles with random orientation are the same as they are for spheres, but values of g_0 are different. Values of g_0 can be larger than those for spherical particles (such as infinite circular cylinders and spheroids at large or small values of shape parameter ξ), smaller than those for spherical particles (hexagonal cylinders and spheroids at intermediate values of ξ), or even equal to

Table 6. Values of ψ, β, and g_0 for Hexagonal Cylinders at $n = 1.333$ and Different Values of the Ratio $\nu = L/l$[a]

ν	ψ	β	g_0
0.2	1.9	4.8	0.9031
0.4	1.8	1.7	0.8607
1	1.6	1.1	0.7847
2	1.6	0.9	0.7601
4	1.8	0.7	0.7971
10	1.9	1.5	0.8442
20	1.9	1.5	0.8665

[a]Here L is the length of a hexagonal cylinder and l is the side of a hexagon's cross section.

those for spherical particles. Asymptotic values for asymmetry parameters of large, nonabsorbing, prolate and oblate spheroids g_0 were tabulated for the first time (see Table 3) to our knowledge.

During this research A. Kokhanovsky (who was on leave from the Institute of Physics in Minsk, Belarus) held a Science and Technology Agency of Japan Fellowship. He thanks the staff members of the Earth Observation Research Center of the National Space Development Agency of Japan (Tokyo) for their encouragement throughout his study. The research by A. Macke was supported by the NASA First International Satellite Cloud Climatology Project.

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